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REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
31 July 1996

3. REPORT TYPE AND DATES COVERED
Final Progress Report

4. TITLE AND SUBTITLE
Probing the carrier recombination dynamics of strained semiconductor quantum heterostructures and nanostructures

5. FUNDING NUMBERS
DAAH04-95-1-0386

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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
ARO 34733.1-MS-RIP

11. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

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13. ABSTRACT *(Maximum 200 words)*
The aim of this research is to examine the structural and optical properties of reduced-dimensionality structures (quantum wires and dots) using novel spatially, spectrally, and temporally resolved electron probes. An optimization of the carrier collection efficiency in quantum wires and dots is of paramount importance in utilizing nanostructures in quantum wire and dot lasers. An extremely powerful and unique high-speed time-resolved cathodoluminescence (CL) system has been developed by Prof. Rich using funds from this DURIP award. This system augments an existing scanning electron microscopy-based CL system which had already yielded important non-time-resolved results for quantum heterostructures and nanostructures. The necessary electronic instrumentation for high-speed electron beam blanking, light detection, and time-correlated single photon counting has been purchased and successfully implemented. The carrier recombination dynamics and collection efficiency in GaAs/AlGaAs quantum dots and InGaP quantum wires are being studied using spatially, spectrally, and temporally resolved cathodoluminescence. Our work in 1996 and beyond will focus on studies of the carrier recombination dynamics and optical properties of quantum wire and box nanostructures and the influence of structural defects on these properties.

14. SUBJECT TERMS
Quantum wires, quantum dots, nanostructures, carrier recombination, time-resolved cathodoluminescence

15. NUMBER IF PAGES

16. PRICE CODE

17. SECURITY CLASSIFICATION OR REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

SCIENTIFIC PERSONNEL WHO IMPLEMENTED DURIP PROJECT:

Principal Investigator: Prof. Daniel H. Rich
Graduate Students: H.T. Lin, K. Rammohan, Y. Tang

LIST OF MANUSCRIPTS **CURRENTLY SUBMITTED** UNDER ARO SPONSORSHIP DURING THIS PERIOD (As of July 31, 1996):

1. D.H. Rich, Y. Tang, and H.T. Lin, Time-resolved cathodoluminescence study of carrier relaxation in strained $(InP)_2/(GaP)_2$ quantum wires, Applied Physics Letters, submitted in July 1996.

2. D.H. Rich, Y. Tang, and H.T. Lin, Linearly polarized and time-resolved cathodoluminescence study of strain-induced laterally ordered $(InP)_2/(GaP)_2$ quantum wires, Physical Review B, submitted in July 1996.

LIST OF MANUSCRIPTS **ACCEPTED FOR PUBLICATION** UNDER ARO SPONSORSHIP DURING THIS PERIOD:

3. D.H. Rich, H.T. Lin, A. Konkar, P. Chen, and A. Madhukar, Time-resolved cathodoluminescence study of carrier relaxation in GaAs/AlGaAs layers grown on a patterned GaAs(001) substrate, Applied Physics Letters, in press (Vol. 69, 1996).

4. H.T. Lin, D.H. Rich, O. Sjölund, M. Ghisoni, and A. Larsson, Cathodoluminescence study of the influence of misfit dislocations on hole accumulation in a *n*-AlGaAs/*p*-GaAs/*n*-InGaAs heterojunction phototransistor, Applied Physics letters, in press (Vol. 69, 1996).

LIST OF EQUIPMENT PURCHASED WITH DURIP FUNDS:

Component	Cost ($)
1) Components for time-correlated single photon counting: Nanosec Delay 425A Gate/Delay Gen. 416A Time-to-Vol. Conv. 457 Pulse Amp. VT110 Discriminator 583 Multi-Channel Analyzer (8k TRUMP Card for Windows) *EG&G ORTEC* *100 Midland Road, Oak Ridge, TN 37831-0895* *Ph:(800) 251-9750 (David Blank)* P.O. # 689113	9,378.79

2) Electrostatic Beam Blanker *Raith USA* *70C Carolyn Blvd., Farmingdale, NY 11735* *Ph: (516) 293-0187 (George Lanzarotta)* P.O. # 688868	25,439.30
3) Pulse Generator AVMP-2-C-USC1 100 ps *Avtech Electrosystems Ltd.* *P.O. Box 5120, Stn. F, Ottawa, Ontario, Canada K2C 3H4* *Ph: (613) 226-5772 (Walter J. Chudobiak)* P.O. # 689572	3,501.89
4) Si Avalanche Photon Counting Module *EG&G Optoelectronics Canada* *22001 Dumberry Road* *Vaudreuil, Quebec* *J7V 8P7, Canada* *Ph:(805) 647-6944 (Mark Nicklas)* P.O. # 13358B	3,875.35
5) 120 MHz Pentium Computer *DELL Computer Corp.* *2214 West Bräker Ln, Bldg. 3* *Austin, TX 78758* *Ph: (800) 727-1100 (Linda Temple)* P.O. # 691970	2,023.19
6) Si CCD array imaging System *Princeton Instruments* *3660 Quaker Bridge Road* *Trenton, NJ 08619* *Ph: (510) 656-9142 (Jay Moscovic)* P.O. # 13360B	21,753.50
7) 0.275 meter spectrograph system *Acton Research Corp.* *P.O. Box 2215* *Acton, MA 01720* *Ph: (508) 263-3584 (Michael Gasek)* P.O. # 13362B	7,107.08
8) Electroformed Custom Copper Mirrors *ORC Electroformed Products* *1300 Optical Drive*	591.05

Azusa, CA 91702
Ph: (818) 969-3344 (Rob Houseman)
P.O. # 002060

9) Stylus color pro 720 dpi ink jet printer 604.04
Sehi Computer Products, Inc.
San Clement, CA 92673
Ph: (800) 233-7344
P.O. # 001031

Total Cost 74,274.19

EXPLANATION OF INSTRUMENTATION PURCHASE:
Items 1-5 comprise the new time-resolved CL system, the basis of the DURIP implementation. Item 5 is a computer that is used to operate the 8k TRUMP card multi-channel analyzer in Item 1. Items 6 and 7 are used for high speed spectra acquisition when the CL system is in the non-time-resolved mode. Item 8 is the mirror used to collect light in all CL modes of operation. Item 9 is a high-resolution color and gray-scale ink-jet printer that is used to obtain hard copies of the CL images.

DESCRIPTION OF RESEARCH PERFORMED WITH EQUIPMENT:

A. Strain-induced laterally ordered $(InP)_2/(GaP)_2$ quantum wire arrays

The optical properties of $(InP)_2/(GaP)_2$ quantum wire arrays were examined with the new time-resolved cathodoluminescence equipment. An In and Ga composition modulation of ~18% forms during the metal-organic chemical-vapor deposition growth of short period $(InP)_2/(GaP)_2$ bilayer superlattices (BSLs). The BSL, owing to the strain-induced lateral ordering (SILO) process, spontaneously orders into quantum wire (QWR) arrays. Transmission electron microscopy showed a period of ~800 Å along the [110] direction, resulting in coherently strained quantum wires. A strong excitation dependence of the polarization anisotropy and energy of excitonic luminescence from the quantum wires was found. The results are consistent with a phase-space and band filling model that is based on a **k·p** and 2D quantum confinement calculation which takes the coherency strain into account. CL images reveal that defects in the quantum wires originate from the GaAs substrate and/or the initial stages of InGaP growth. The effects of defects on the band filling, carrier relaxation kinetics, and nonlinear optical properties were examined.

Prof. Rich and his students have performed a detailed study of the band-filling and polarization anisotropy of $(InP)_2/(GaP)_2$ QWRs using time-resolved CL and linearly polarized CL. Calculations of the excitation-dependent polarization anisotropy and CL energy shifts with a **k·p** model that incorporates both strain and quantum confinement were performed. The calculations confirm a strong interplay between band-filling and e-h wavefunction overlap, which leads to very interesting and possibly useful nonlinear optical and polarization properties. The linearly polarized CL and excitation-dependent CL data are consistent with the calculations.

Time-resolved CL measurements show a rapid capture of carriers from the $In_{0.49}Ga_{0.51}P$ barrier on a scale of less than ~ 100 ps. Time-delayed spectra have been examined to connect changes in CL linewidth and position with band-filling in the QWR during the *onset* and *decay* of the electron beam pulses. Experiments performed at various temperatures show that thermal activation of carriers in the QWR and transfer to and from the $In_{0.49}Ga_{0.51}P$ barriers plays an important role in determining the measured lifetimes. A decrease in lifetime at higher temperatures is found to be caused likely by thermal re-emission from the QWR to the barrier.

The optical properties and crystal quality of the $In_{0.49}Ga_{0.51}P$ barriers and $(InP)_2/(GaP)_2$ BSL strongly depended on the growth and sample surface conditions. Samples exhibiting nonuniform bright and dark regions in the CL imaging show large spatial variations in the phase-space filling and nonlinear optical properties. These defects are associated with (i) the starting GaAs substrate or (ii) the initial stages of $In_{0.49}Ga_{0.51}P$ growth on GaAs in which In-rich regions at the GaAs surface form. The subsequent growth of a $(InP)_2/(GaP)_2$ BSL over the $In_{0.49}Ga_{0.51}P$ buffer layer resulted in SILO QWRs. CL imaging showed that dark-defect regions arising in the GaAs, QWR, and $In_{0.49}Ga_{0.51}P$ emissions correlate spatially. A narrow luminescence lineshape for the QWR emission was observed for a sample exhibiting a high degree of homogeneity in the CL imaging.

Two samples exhibiting disorder in the CL imaging show a broadening of the QWR luminescence lineshape into two or more components. A detailed study of the excitation-dependence and spatial correlations of these components has been performed for a BSL sample which exhibits two QWR-like components in its CL spectrum. Time-resolved CL and excitation-dependent CL experiments show that diffusive transport and thermalization of carriers occur between two distinct QWR regions giving rise to the two components comprising the QWR spectral lineshape. CL wavelength imaging shows that defects existing in the barrier reduce the sensitivity of the QWR to band-filling, owing to the enhancement of the nonradiative recombination rate and local reduction of the steady-state carrier density. In conclusion, these results demonstrate the need to minimize the defect density and maximize the homogeneity in SILO QWR samples in order to optimize the nonlinear optical effects associated with phase-space filling.

The complete results are present in papers 1 and 2, currently submitted to Applied Physics Letters and Physical Review B, respectively.

B. Carrier relaxation in 3D confined GaAs/AlGaAs layers grown on patterned GaAs(001) substrates

The growth of $GaAs/Al_xGa_{1-x}As$ nanostructures (quantum wires and boxes) on patterned GaAs substrates has attracted much attention in recent years. The high quality of the hetero-layers and interfaces that can be achieved during a single growth step circumvents the need for post-growth etching and patterning on a nanoscale that can inevitably degrade the optical and structural quality of the nanostructure and its surrounding barriers. As a result of facet-dependent growth and cation migration rates, growth on pre-patterned substrates can be used to attain a size reduction in the as-patterned μm-scale GaAs mesas so that the resulting size of three-dimensionally confined regions are on the order of ~ 100 Å. From a fundamental perspective,

the study of quantum boxes fabricated on isolated mesas should permit a simple understanding of the temperature and excitation dependence of its optical properties. Such isolated nanostructures on regular arrays inherently avoid a more complex environment of thermally excited electrons and holes that can potentially interact with several quantum boxes, as in the case for the self-organized InAs island growth. A basic issue that needs to be addressed with the growth of nanostructures on mesas concerns the tailoring of the structure in a manner that maximizes carrier capture and collection into the quantum box. Studies of carrier thermalization and collection have been performed for GaAs/$Al_xGa_{1-x}As$ quantum wires grown into V-shaped grooves. From a device viewpoint, the efficiency of carrier collection in the nanostructure determines the quantum efficiency of the quantum wire or box laser.

Prof. Rich and his students have examined the kinetics of carrier relaxation in three-dimensionally (3D) confined GaAs/AlGaAs layers obtained by growth on pre-patterned GaAs(001) with time-resolved cathodoluminescence (CL). Time-delayed CL spectra at 87 K reveal that *(i)* relaxation of hot carriers into the largest 3D confined regions occurs on a time scale of a few hundred ps during the onset of luminescence, and *(ii)* the luminescence decay time also increases for these larger confined regions, owing to thermal re-emission from QWs, diffusion across $Al_xGa_{1-x}As$ barriers, and carrier feeding from surrounding thinner QWs. The samples were prepared in collaboration with Prof. Anupam Madhukar at USC. *This DURIP enabled, again, the development of the time-resolved CL system from which data on the carrier relaxation dynamics was obtained.*

The complete results are present in paper 3, appearing in Applied Physics Letters.

C. Influence of misfit dislocations on hole accumulation in a *n*-AlGaAs/*p*-GaAs/*n*-InGaAs heterojunction phototransistor

We have studied the influence of misfit dislocations on hole accumulation in the base layer of a *n*-AlGaAs/*p*-GaAs/*n*-InGaAs heterojunction phototransistor (HPT). Spatially and time-resolved CL measurements reveal that variations in the hole accumulation is caused primarily by strain-induced defects which impede the transport of holes in the collector. The lifetime of holes in the InGaAs/GaAs collector is found to be negligibly affected by the underlying misfit dislocations in the InGaAs/GaAs collector. The reduction in the local electron-beam-induced current signal by the dislocations is less than $\sim 20\%$, indicating that these defects have a minor impact on the overall device performance. The samples were supplied by Prof. Anders Larsson of Chalmers University in Sweden.

The complete results are present in paper 4, appearing in Applied Physics Letters.

IMPACT OF THIS DURIP AWARD ON FUTURE RESEARCH:

This award has enabled the development of a time-resolved CL system and augments the spatially and spectrally resolved capabilities that existed prior to the award. Together with other recent innovations of our system, including the electron beam-induced absorption and

polarization detection capabilities, this DURIP award has facilitated the development of one of the most unique optical microprobes in the world today. The DURIP implementation has already enabled the gleaning of important information concerning the carrier relaxation dynamics of quantum nanostructures, as discussed above. Students involved in this project have and will continue to benefit from exposure to these extremely high-profile and innovation optical microcharacterization probes. We anticipate that many more studies on the carrier relaxation dynamics of quantum heterostructures and nanostructures, prepared using a variety of growth and fabrication techniques, will soon commence. From these studies we hope to gain better insights into the physics of carrier collection and relaxation in nanostructures. For example, we hope to better understand what barriers impede transport to and collection into the quantum wire or box. Such issues are paramount in maximizing the quantum efficiency of quantum wire or box lasers. The time-resolved experiments provide critical snap-shots of the carrier recombination and luminescence lineshapes. The above results and papers are expected to serve as archetypes for the type of future experiments that will be performed in Prof. Rich's laboratory. In short, the objectives of the ARO-DURIP proposal are completely fulfilled. Prof. Rich and students have, in less than a year's time established a viable program, bearing fruit in the form of published results and conference presentations.